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(54) RESIN COMPOSITION AND ITS PREPARATION

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a resin compsn. which contains a filler or layered silicate having a small particle size and homogeneously dispersed therein by mixing a thermoplastic resin with an inorg. filler under contact with a supercritical fluid.

SOLUTION: This resin compsn. is prepd. by mixing 100 pts.wt. thermoplastic resin with an inorg. filler in an amt. of 80-0.01 wt.% of the compsn. and 0.05-10 pts.wt. olefin compd. having a carboxylic anhydride group under contact with carbon dioxide or the like which becomes a supercritical fluid at a temp. in the range from the m.p. of the resin to its decomposition temp. in an amt. of the fluid of 0.01-30 wt.% of the compsn. Examples of the thermoplastic resin are a polyamide resin, a polyester resin, and a polyarylene sulfide resin. The inorg. filler is a fibrous one (e.g. glass fibres or steel fibres), a powdery or platy one (e.g. tale or mica), or a layered silicate (e.g. montmorillonite), and 0.01-20 wt.% coupling agent (e.g. a silane compd.) is added to the filler.

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CLAIMS

[Claim(s)]

[Claim 1] (A) The resin constituent which thermoplastics and (B) inorganic filler are made to contact (C) supercritical fluid, and comes to knead them.

[Claim 2] (B) Fat constituent according to claim 1 whose content of an inorganic filler is 80 - 0.01 % of the weight among a constituent.

[Claim 3] (A) The resin constituent according to claim 1 or 2 characterized by thermoplastics being one or more sorts of resins chosen from polyamide resin, a saturated-polyester resin, ABS plastics, and the polyphenylene-sulfide resin.

[Claim 4] (B) Resin constituent according to claim 1 to 3 whose inorganic filler is a silicic acid system inorganic filler.

[Claim 5] (C) The resin constituent according to claim 1 to 4 whose supercritical fluid is a super-criticality carbon dioxide.

[Claim 6] (A) making thermoplastics and (B) inorganic filler contact (C) supercritical fluid, and kneading them in a batch-type mixing chamber, -- claims 1-4 -- the manufacture method of the resin constituent characterized by manufacturing the resin constituent of a publication to either

[Claim 7] (A) making thermoplastics and (B) inorganic filler contact (C) supercritical fluid, and kneading them, using the mixer of continuous system, -- claims 1-4 -- the manufacture method of the resin constituent characterized by manufacturing the resin constituent of a publication to either

[Claim 8] (A) taking out out of a system by making (C) supercritical fluid into a gas by reducing the pressure in a system, when thermoplastics and (B) inorganic filler are made to contact (C) supercritical fluid, and are kneaded and kneading is completed -- claims 5 and 6 -- the manufacture method of the resin constituent characterized by manufacturing the resin constituent of a publication to either

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] It distributes without an inorganic filler condensing in thermoplastics, and this invention has the outstanding mechanical and thermal property, and relates to the manufacture method at the resin constituent row which is excellent in the appearance of mold goods.

[0002]

[Description of the Prior Art] By adding an inorganic filler to thermoplastics, raising the intensity, rigidity, and thermal resistance is known well. However, when the particle diameter of the inorganic filler to be used is very small, particles condense, distribution in thermoplastics becomes poor, and there are problems -- shock resistance is spoiled. Moreover, differential powder of the sheet silicate is carried out to nano meter order into the polyamide which is thermoplastic engineering plastics, and the mechanical strength and the attempt which improves rigidity are performed in recent years. in this clay-polyamide complex, although rigidity is improved sharply, for that, a sheet silicate (clay) calls it about 1nm in thickness -- it is necessary to distribute in a polyamide in the state of a monolayer mostly. However, usually, a sheet silicate and thermoplastics were mixed and kneaded, secondary condensation arose in the request, and uniform distribution into a resin was difficult. Although examination which is going to obtain uniform distribution by using the intercalation compound which makes a sheet silicate a host at JP,8-12881,A, and makes specific quarternary ammonium salt a guest is made, improvement in much more mechanical property is called for. Moreover, the thing for which the vent-port which carried out melting kneading with the resin after making JP,8-151449,A and JP,9-48856,A swell a clay mineral with a solvent, and was prepared in the extruder is held to reduced pressure, Although the attempt which is going to obtain uniform distribution by removing a solvent was indicated, in order to use a solvent, when there was a problem that the process becomes complicated, the usual solvent had the problem that a thing with the operation which swells a sheet silicate did not have sufficient operation to thermoplastics, either.

[0003]

[Problem(s) to be Solved by the Invention] Then, this invention makes it a technical problem to obtain the resin constituent which it makes it come uniformly to distribute, without making the small filler and small sheet silicate of solving an above-mentioned problem, i.e., an inorganic filler division particle diameter, condense in thermoplastics.

[0004]

[Means for Solving the Problem] Then, the result which repeated examination wholeheartedly in order that this invention persons might solve an above-mentioned problem, By making low-molecular [low-molecular / of the super-critical state], i.e., supercritical fluid, act, in case thermoplastics is made to distribute the small filler and small sheet silicate of an inorganic filler division particle size. On the usual kneading conditions, the uniform distributed state which cannot be acquired because of secondary condensation could be attained, and it found out that the resin constituent which has the mechanical property which was excellent as the result was obtained, and resulted in this invention.

[0005] That is, this inventions are the resin constituent which (A) thermoplastics and (B) inorganic filler are made to contact (C) supercritical fluid, and comes to knead them, and its manufacture method.

[0006]

[Embodiments of the Invention] this invention is explained in detail below.

[0007] If (A) thermoplastics in this invention is a resin which can carry out plastic working by heating, either can be used regardless of amorphia, crystallinity, and mesomorphism. (A) As a desirable example of thermoplastics, polyester resin, polyamide resin, the poly arylene sulfide resin, polyacetal resin, ABS plastics, a polyphenylene-oxide resin, polycarbonate resin, polystyrene resin, polyolefin resin, acrylic resin, etc. are mentioned, and two or more sorts of these may be used together. Especially, polyester resin, polyamide resin, and the poly arylene sulfide resin can be used preferably.

[0008] With polyester resin here, the thermoplastic polyester which has a ring per chain of a polymer is mentioned, and the polymer or copolymer obtained by the condensation reaction which makes a principal component an aromatic dicarboxylic acid (or the ester formation nature derivative), a diol (or the ester formation nature derivative), and/or hydroxycarboxylic acid is usually specifically mentioned. Even if these are the things of mesomorphism, they may be the things of non-mesomorphism.

[0009] In this invention as an example of desirable polyester resin As a thing of non-mesomorphism, a polyethylene terephthalate, polypropylene terephthalate, To a polybutylene terephthalate and Pori, xylene terephthalate, polyethylene -2, 6-naphthalene dicarboxy rate, Others [rate / 4'-dicarboxy / a polybutylene -2, 6-naphthalene dicarboxy rate, polyethylene -1, 2-screw (phenoxy)

ethane -4, and]. Polyethylene isophthalate / terephthalate, polybutylene isophthalate / terephthalate, a polybutylene terephthalate / Decan dicarboxy rate, polyethylene -4, a 4'-dicarboxy rate / terephthalate, etc. are mentioned.

[0010] The polyester which forms the anisotropy melting phase which consists of a structural unit chosen from the aromatic oxy-carbonyl unit, the aromatic dioxy unit, the aromatic dicarbonyl unit, the ethylene dioxy unit, etc. as a thing of mesomorphism can be mentioned.

[0011] As an aromatic oxy-carbonyl unit, for example as the structural unit generated from para hydroxybenzoic acid and the 6-hydroxy-2-naphthoic acid, and an aromatic dioxy unit for example, as the structural unit generated from 4, 4'-dihydroxy biphenyl, hydroquinone, or t-butyl hydroquinone, and an aromatic dicarbonyl unit For example, as a terephthalic acid, an isophthalic acid, 2, the structural unit generated from 6-naphthalene dicarboxylic acid, and an aromatic imino OKISHI unit, the structural unit generated from 4-aminophenol is mentioned, for example.

[0012] It is also suitable practically to divide as polyester resin, and for a polybutylene terephthalate, a polyethylene terephthalate, polyethylene -2, and 6-naphthalene dicarboxy rate to be mentioned as a desirable thing, and to use these polyester resin as mixture according to required properties, such as a moldability, thermal resistance, toughness, and front-face nature.

[0013] Polyamide resin is a polyamide which uses amino acid, a lactam or a diamine, and a dicarboxylic acid as a main raw material. As an example of representation of the raw material, 6-aminocaproic acid, a 11-amino undecanoic acid, Amino acid, such as 12-amino dodecanoic acid and an paraamino methyl benzoic acid, Lactams, such as an epsilon caprolactam and omega-RAURO lactam, a tetramethylenediamine, A hexamethylenediamine, 2-methyl pentamethylene diamine, an undecamethylene diamine, A dodeca methylene diamine, 2 and 2, 4-/4, a 4-trimethyl hexamethylenediamine, [2 and 4] 5-methyl nonamethylene diamine, a meta-xylene diamine, a PARAKI silylene diamine, 1, 3-screw (aminomethyl) cyclohexane, 1, 4-screw (aminomethyl) cyclohexane, 1-amino-3-aminomethyl - 3, 5, and 5-trimethyl cyclohexane, Screw (4-amino cyclohexyl) methane, screw (3-methyl-4-amino cyclohexyl) methane, 2 and 2-screw (4-amino cyclohexyl) propane, a screw (aminopropyl) piperazine, Aliphatic series, such as an aminoethyl piperazine, an alieycle group, an aromatic diamine, And an adipic acid, a SUPERIN acid, an azelaic acid, a sebacic acid, dodecane 2 acid, A terephthalic acid, an isophthalic acid, 2-chloro terephthalic acid, 2-methyl terephthalic acid, 5-methyl isophthalic acid, 5-sodium sulfoisophtharate, a hexahydro terephthalic acid, Aliphatic series, such as a hexahydro isophthalic acid, an alieycle group, and an aromatic dicarboxylic acid are mentioned, and the polyamide homopolymer or copolymer guided from these raw materials can be respectively used in the form of independent or mixture in this invention.

[0014] In this invention especially useful polyamide resin It is polyamide resin excellent in the thermal resistance which has the melting point 200 degrees C or more, or intensity. As a concrete example, a PORIKA pro amide (nylon 6), a polyhexamethylene adipamide (Nylon 66), Polytetra ethylene adipamide (Nylon 46), a polyhexamethylene sebacamide (Nylon 610), Polyhexamethylene DODEKAMIDO (Nylon 612), a polyhexamethylene adipamide / polyhexamethylene terephthalamide copolymer (Nylon 66 / 6T), A polyhexamethylene adipamide / polyhexamethyleneisophthalamide copolymer (Nylon 66 / 6I), A polyhexamethylene adipamide / polyhexamethylene terephthalamide / polyhexamethyleneisophthalamide copolymer (Nylon 66 / 6T/6I), poly xylylene adipamide (nylon XD6) and such mixture, or a copolymer is mentioned. It is also suitable practically to be able to give examples, such as nylon 6, Nylon 66, Nylon 610, nylon 6/66 copolymer, and nylon 6/12 copolymer, and to use these Nylons as mixture as an especially desirable thing, according to required properties, such as a moldability, thermal resistance, toughness, and front-face nature, further.

[0015] There is no limit especially in the polymerization degree of these Nylons, and the thing of the range of 1.5-5.0, especially the range of 2.0-4.0 has the desirable relative viscosity measured at 25 degrees C among 1% of concentrated-sulfuric-acid solution.

[0016] As a poly arylene sulfide resin, it can use both a bridge formation type a straight chain type and branching type.

[0017] with (B) inorganic filler used for this invention, it is usually used as the filler of a resin, and a reinforcement -- fibrous or things [being un-fibrous] (for example, a grain, powder, or a tabular etc.) are pointed out As a desirable example of a fibrous inorganic filler, a glass fiber, a potassium titanate fiber, plaster fiber, brass fiber, stainless steel fiber, copper fiber, steel fiber, boron whisker fiber, a carbon fiber, etc. are mentioned. moreover, as a desirable example of the inorganic filler of a grain, powder, and a tabular A WARASUTE night, a sericite, a kaolin, a mica, clay, a bentonite, Silicate, such as talc and an alumina silicate, an alumina, silicon oxide, Metallic oxides, such as a magnesium oxide, a zirconium oxide, and titanium oxide, Carbonates, such as a calcium carbonate, a magnesium carbonate, and a dolomite, a calcium sulfate, Sulfates, such as a barium sulfate, a glass bead, a glass milled fiber, boron nitride, silicon carbide, Saroyan, etc. may be mentioned, and these may be hollow (for example, a hollow glass fiber, a glass micro balloon, a milt balloon, a carbon balloon, etc.). Although according to this invention the inorganic filler illustrated above is uniformly distributed in thermoplastics and the improvement effect of a mechanical property is accepted as a result, the effect of especially distributing the inorganic filler of a particle system uniformly is large. a thing with a fibrous particle system inorganic filler here -- the diameter -- about a grain and a powdery thing, the minor axis says [thing / of a tabular / the thickness] a thing 1 micrometer or less on an average As an inorganic filler with which the technology of this invention appears most notably, there is a silicate system inorganic filler and the effect over a sheet silicate is large also in especially inside. As an example of a sheet silicate, bloating tendency synthetic micas, such as various clay minerals, such as smectite system clay minerals, such as a montmorillonite, a beidellite, a nontronite, a saponite, a hectorite, a sauconite, and a vermiculite, a halloysite, a money dynamite, KENIYAITO, a zirconium phosphate, and phosphoric acid titanium, Li type fluorine TENIO light, Na type fluorine TENIO light, a Na type 4 silicon fluorine mica, and a Li type 4 silicon fluorine mica, etc. are mentioned, and it may be compounded even if it is a Bloating tendency synthetic micas, such as smectite system clay minerals, such as a montmorillonite and a hectorite, and a Na type 4 silicon fluorine mica, Li type fluorine TENIO light, are desirable also

in these. Although special processing is not necessarily needed when using a sheet silicate as a (B) inorganic filler in this invention, when a sheet silicate has the cation of convertibility between layers, the convertibility cation can use preferably the sheet silicate exchanged by the organic onium ion. In these, an ammonium ion and phosphonium ion are desirable and especially an ammonium ion is used preferably. As an ammonium ion, any of the 1st class ammonium, the 2nd class ammonium, the 3rd class ammonium, and the 4th class ammonium are sufficient.

[0018] As the 1st class ammonium ion, desyl ammonium, dodecyl ammonium, octadecyl ammonium, oleyl ammonium, benzyl ammonium, etc. are mentioned

[0019] As the 2nd class ammonium ion, methyl dodecyl ammonium, methyl octadecyl ammonium, etc. are mentioned

[0020] Dimethyl dodecyl ammonium, dimethyloctadecyl ammonium, etc. are mentioned as the 3rd class ammonium ion.

[0021] As the 4th class ammonium ion, benzyl trimethylammonium, Benzyl triethyl ammonium, benzyl tributyl ammonium, Benzyl trialkyl ammonium ions, such as benzyl dimethyl dodecyl ammonium and benzyl dimethyloctadecyl ammonium, Trioctyl methylammonium, trimethyl octyl ammonium, Alkyl trimethyl ammonium ions, such as trimethyl dodecyl ammonium and trimethyl octadecyl ammonium, Dimethyl dialkyl ammonium ions, such as dimethyl dioctyl ammonium, dimethyl didodecyl ammonium, and dimethyl dioctadecyl ammonium, etc. are mentioned.

[0022] Moreover, the ammonium ion guided besides these from an aniline, p-phenylene diamine, alpha-naphthylamine, a p-amino dimethylaniline, a benzidine, a pyridine, a piperidine, 6-aminocaproic acid, a 11-amino undecanoic acid, 12-amino dodecanoic acid, etc. is mentioned.

[0023] Also in these ammonium ions, the sum total of the carbon number in the molecule of an ammonium ion is suitable for especially the 4th class ammonium ion of 11-30. Specifically, they are trioctyl methylammonium, trimethyl octadecyl ammonium, benzyl dimethyl dodecyl ammonium, benzyl dimethyloctadecyl ammonium, etc.

[0024] Moreover, it is desirable to use the (B) inorganic bulking agent by the coupling agents (for example, an isocyanate system compound, an organic silane system compound, an organic titanate compound, an organic borane system compound, an epoxy compound, etc.) which have a reactant functional group, carrying out preliminary processing in the meaning which obtains the more excellent mechanical strength. Especially a desirable coupling agent is an organic silane system compound (silane coupling agent), as the example Gamma-glycidoxypolytrimethoxysilane, gamma-glycidoxy propyl TORIETOKISHISII silane, Epoxy-group content alkoxysilane compounds, such as beta-(3, 4-epoxycyclohexyl) ethyl trimethoxysilane, Sulfhydryl-group content alkoxysilane compounds, such as gamma-mercapto propyltrimethoxysilane and gamma-mercapto propyl triethoxysilane, gamma-ureido propyl triethoxysilane, gamma-ureido propyl trimethoxy SHISHIRAN, Ureido machine content alkoxysilane compounds, such as gamma-(2-ureido ethyl) aminopropyl trimethoxysilane, gamma-isocyanato propyl triethoxysilane, gamma-isocyanato propyltrimethoxysilane, gamma-isocyanato propylmethyl dimethoxysilane, gamma-isocyanato propylmethyl diethoxysilane, gamma-isocyanatopropylethyl dimethoxysilane, gamma-isocyanatopropylethyl diethoxysilane, Isocyanato-group content alkoxysilane compounds, such as gamma-isocyanato propyl trichlorosilane, gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, Amino-group content alkoxysilane compounds, such as gamma-(2-aminoethyl) aminopropyl trimethoxysilane and gamma-aminopropyl trimethoxysilane, Hydroxyl-group content alkoxysilane compounds, such as gamma-hydroxypropyl trimethoxysilane and gamma-hydroxypropyl triethoxysilane, Carbon carbon unsaturation machine content alkoxysilane compounds, such as gamma-methacryloxypropyl trimethoxy silane, vinyltrimethoxysilane, and N-beta-(N-vinylbenzyl aminoethyl)-gamma-aminopropyl trimethoxysilane, a hydrochloride, etc. are mentioned. Especially, gamma-methacryloxypropyl trimethoxy silane, gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, gamma-(2-aminoethyl) aminopropyl trimethoxysilane, and gamma-aminopropyl trimethoxysilane are used preferably. Although the method of these silane coupling agents carrying out surface treatment of the (B) inorganic bulking agent beforehand according to a conventional method, and subsequently carrying out (A) thermoplastics and (C) melting kneading is used preferably, in case melting kneading of (A) thermoplastics and the (B) inorganic bulking agent is carried out under existence of low-molecular [of the (C) super-critical state], without performing surface treatment of an inorganic bulking agent beforehand, you may use the so-called integral blending method which adds these coupling agents.

[0025] Usually, although a coupling agent is used to an inorganic bulking agent in about 0.01 - 20% of the weight of the range, its 0.05 - 15 % of the weight is desirable.

[0026] As for (B) inorganic filler in this invention, it is desirable from the point of a moldability to blend preferably, 80 to 0.01% of the weight, by the inorganic ash content in a constituent, so that it may become 50 - 0.1 % of the weight. In addition, the inorganic ash content in a constituent is the value which was made to ash 2g of constituents for 3 hours, and asked for them with the 500-degree C electric furnace.

[0027] It is also effective to add the polymer of the olefin compound which has a carboxylic-acid anhydride machine in a molecule if needed in this invention, or these olefins compound at the time of kneading to the purpose which improves a mechanical property. As an example of the olefin compound which has a carboxylic-acid anhydride machine in a molecule, or these olefins compound, the polymer of a maleic anhydride, itaconic acid anhydride, an anhydrous glutaric acid, an anhydrous citraconic acid, an anhydrous aconitic acid, or these substitution olefin compound etc. is mentioned. In addition, olefins other than the olefin compound which has carboxylic-acid anhydride machines, such as styrene, an isobutylene, a methacrylic ester, and an acrylic ester, in a molecule in the polymer of an olefin compound are books. As for the polymerization degree of the polymer of an olefin compound, 2-100 are desirable, 2-50 are more desirable, and 2-20 are the most desirable to a pan. In these, a maleic anhydride and the poly maleic anhydride are used most preferably. As a poly maleic anhydride, the thing of a publication can be used for J.Macromol. Sci.-Revs.Macromol.Chem., C13 (2), 235 (1975), etc., for example.

[0028] The addition in the case of adding the polymer of the olefin compound which has these carboxylic-acids anhydride machine in a molecule, or these olefins compound has desirable 0.05 - 10 weight section from the point of the fluidity of the improvement effect of an impact strength, and a constituent to the (A) thermoplastics 100 weight section, it is desirable still more desirable that it is the range of further 0.1 - 5 weight sections, and it is 0.1 - 3 weight section.

[0029] The resin constituent of this invention is obtained by making (A) thermoplastics and (B) inorganic filler contact (C) supercritical fluid, and kneading them. As a mode of kneading, melting kneading is desirable.

[0030] Supercritical fluid here is the 3rd fluid which is not the any, either, although it has the middle-property of a gas and a liquid in the state more than the critical pressure more than critical temperature, and it has the big diffusion coefficient compared with the liquid. As matter which can make supercritical fluid comparatively easily, although a carbon dioxide, ammonia, methane, etc. are mentioned, in order to use it in this invention, what may serve as supercritical fluid in the temperature field below a decomposition temperature is usually desirable more than the melting temperature of thermoplastics. Moreover, as supercritical fluid used for this invention, it is the point of an effect, and a carbon dioxide is the most desirable.

[0031] The amount of the supercritical fluid used in this invention has 0.01 - 30 desirable % of the weight to the constituent made to contact, and its 0.1 - 20 % of the weight is especially desirable.

[0032] In this invention, although especially restrictions do not have equipment for making (A) thermoplastics and (B) inorganic filler contact (C) supercritical fluid, and kneading them, it is more than the melting working temperature of (A) thermoplastics, and if it is equipment which can be pressurized more than the critical pressure, even if it is any of a batch type and continuous system, it can be used preferably. As an example, what prepared the inlet of a pressurization fluid can be preferably used for the extruder of the monopodium used for melting kneading of thermoplastics, and two shafts. It is effective to form the equipment for discharging out of a system by making low-molecular [of the super-critical state] into a gas by preparing the inlet which introduces supercritical fluid in the position where thermoplastics will be in the state where it fused completely from the plasticization zone of an extruder by the nozzle side, and decompressing in more detail, the vent-port prepared in the position which kneading ends completely.

[0033] Furthermore, to the resin constituent of this invention, additives, such as release agents, such as antioxidants, such as various addition components in ordinary use, a crystalline-nucleus agent, a coloring inhibitor, a hindered phenol, and hindered amine, ethylene-bis-stearylamine, and higher-fatty-acid ester, a plasticizer, a thermostabilizer, lubricant, an ultraviolet-rays inhibitor, a coloring agent, and a flame retarder, can be added in the range which does not spoil the purpose of this invention.

[0034] Let the thermoplastics constituent of this invention be mold goods easily by the usual processing methods, such as extrusion molding and injection molding.

[0035]

[Example] Although an example is shown below and this invention is explained to it still more concretely, this invention is not limited to the publication of these examples. Moreover, all the blending ratio of coal is the weight sections.

[0036] 30mm twin screw extruder which has a vent-port into the portion from a gas addition mouth and the delivery of a kneading zone is used for the portion from the resin feed hopper of the kneading zone set as 1250 degrees C of examples. The kaolin 15 weight section by which silane-coupling-agent processing of 1% of concentration and the relative viscosity measured at 25 degrees C is carried out with 85 weight sections in the nylon 6 of 2.74 is continuously supplied among a concentrated sulfuric acid. While maintaining [a liquefaction carbon-dioxide bomb] the pressure more than the critical pressure at the gas addition mouth furthermore prepared in an extruder barrel for about 3% of the weight of a carbon dioxide to a tie in and a constituent through high pressure pumping. It introduced and the vent-port was further changed into the reduced pressure state, and it kneaded, deaerating, and the pellet was obtained. Injection molding was carried out after drying the obtained constituent pellet at 250 degrees C of cylinder temperatures, and 80 degrees C of die temperatures, and the cylindrical test piece of thickness 1 / x1 / 8 "ASTM No. 1 test piece and 1/2" x5"4" ** was fabricated.

[0037] An ASTM No. 1 test piece is used and it is ASTM. According to the D638 method, a cylindrical test piece is used for a tension test again, and it is ASTM. The result shown in a bending test deed and Table 1 according to the D790 method was obtained.

[0038] Example of reference 1Na type montmorillonite (Kunimine Industries : KUNIPIAF, cation exchange capacity 120 mEq / 100g) 100g was scattered to 10l. of warm water a stirred part, warm water 2l. which dissolved trioctylmethylammonium chloride 48g (cation exchange capacity and equivalence) here was added, and it stirred for 1 hour. Warm water washed, after carrying out produced precipitation a ** exception, this washing and ** -- the organic-ized sheet silicate which performed another operation 3 times, carried out the vacuum drying of the obtained solid-state at 80 degrees C, and was dried was obtained When the inorganic ash content of the obtained organic-ized sheet silicate was measured, the rate of inorganic ash content was 67 % of the weight. In addition, the inorganic ash content of a sheet silicate is the value which ashed 0.1g of sheet silicates for 3 hours, and asked for them with the 500-degree C electric furnace.

[0039] Melting kneading was carried out introducing a super-criticality carbon dioxide at about 2% of the weight of a rate to a constituent using the twin screw extruder which used the cylinder temperature in the example 1 set as 250 degrees C, after example 2 relative viscosity blended the organic-ized sheet silicate 3.8 weight section obtained in the 96.2 weight sections and the example 1 of reference in the nylon 6 of 2.74 and pre blended by the tumbler mixer, and the resin constituent was obtained. After pelletizing the obtained constituent, the vacuum drying of it was carried out at 80 degrees C for 10 hours, it performed injection molding at 250 degrees C of cylinder temperatures, and 80 degrees C of die temperatures, and obtained the test piece. When about 2g of test pieces was made to ash with a 500-degree C electric furnace for 3 hours and the inorganic ash content was

calculated, the rate of inorganic ash content was 2.5 % of the weight. The evaluation result of machine physical properties was shown in Table 1.

[0040] It was used without processing especially Na type montmorillonite used in the example 1 of example 3 reference. Melting kneading was carried out introducing a super-criticality carbon dioxide at about 2% of the weight of a rate to a constituent using the twin screw extruder which used the cylinder temperature in the example 1 set as 250 degrees C, after pre blending the 97.5 weight sections and the Na type montmorillonite 2.5 weight section for the nylon 6 used in the example 1 by the tumbler mixer, and the resin constituent was obtained. After pelletizing the obtained constituent, the vacuum drying of it was carried out at 80 degrees C for 10 hours, it performed injection molding at 250 degrees C of cylinder temperatures, and 80 degrees C of die temperatures, and obtained the test piece.

[0041] After 0.5% of concentration and the intrinsic viscosity measured at 25 degrees C blend the polybutylene-terephthalate 96.2 weight section of 1.2, and the organic-ized sheet silicate 3.8 weight section obtained in the example 1 of reference and pre blends by the tumbler mixer among an example 4 o-chlorophenol solution, while introducing a super-criticality carbon dioxide at about 2% of the weight of a rate to a constituent using the twin screw extruder which used the cylinder temperature in the example 1 set as 250 degrees C. Melting kneading was carried out and the resin constituent was obtained. After pelletizing the obtained constituent, hot air drying of it was carried out at 130 degrees C for 4 hours, it performed injection molding at 250 degrees C of cylinder temperatures, and 80 degrees C of die temperatures, and obtained the test piece. It was 2.5wt%, when about 2g of test pieces was made to ash with a 500-degree C electric furnace for 3 hours and the inorganic ash content was calculated. The evaluation result of machine physical properties was shown in Table 1.

[0042] The same constituent as example of comparison 1 example 1 was created under the nonexistence of the carbon dioxide of the (C) super-critical state. The measurement result of a mechanical property is shown in Table 1.

[0043] The same constituent as example of comparison 2 example 2 was created under the nonexistence of the carbon dioxide of the (C) super-critical state. The measurement result of a mechanical property is shown in Table 1.

[0044] The same constituent as example of comparison 3 example 3 was created under the nonexistence of the carbon dioxide of the (C) super-critical state. The measurement result of a mechanical property is shown in Table 1.

[0045] The same constituent as example of comparison 4 example 4 was created under the nonexistence of the carbon dioxide of the (C) super-critical state. The measurement result of a mechanical property is shown in Table 1.

[0046]

[Table 1]

実施例 3	実施例 4	比較例 1	比較例 2	比較例 3	比較例 4
ナイロン 6	PBT	ナイロン 6	ナイロン 6	ナイロン 6	PBT
有機化層状珪酸塩	有機化層状珪酸塩	カオリン	有機化層状珪酸塩	未処理層状珪酸塩	有機化層状珪酸塩
2.5	2.5	15	2.5	2.5	2.5
2	2	なし	なし	なし	なし
3.28	3.22	4.01	3.23	2.91	3.01
7	6	4	4	4	3

表1	実施例	比較例	平均粒径 (nm)	分散性 (nm)	分散性 (nm)
実施例 (A)	7/2/6	7/2/6	+		
実施例 (B)	7/2/6	7/2/6	+		
実施例 (C)	7/2/6	7/2/6	+		
実施例 (D)	7/2/6	7/2/6	+		
実施例 (E)	7/2/6	7/2/6	+		
実施例 (F)	7/2/6	7/2/6	+		
実施例 (G)	7/2/6	7/2/6	+		
実施例 (H)	7/2/6	7/2/6	+		
実施例 (I)	7/2/6	7/2/6	+		
実施例 (J)	7/2/6	7/2/6	+		
実施例 (K)	7/2/6	7/2/6	+		
実施例 (L)	7/2/6	7/2/6	+		
実施例 (M)	7/2/6	7/2/6	+		
実施例 (N)	7/2/6	7/2/6	+		
実施例 (O)	7/2/6	7/2/6	+		
実施例 (P)	7/2/6	7/2/6	+		
実施例 (Q)	7/2/6	7/2/6	+		
実施例 (R)	7/2/6	7/2/6	+		
実施例 (S)	7/2/6	7/2/6	+		
実施例 (T)	7/2/6	7/2/6	+		
実施例 (U)	7/2/6	7/2/6	+		
実施例 (V)	7/2/6	7/2/6	+		
実施例 (W)	7/2/6	7/2/6	+		
実施例 (X)	7/2/6	7/2/6	+		
実施例 (Y)	7/2/6	7/2/6	+		
実施例 (Z)	7/2/6	7/2/6	+		

[0047]

[Effect of the Invention] The resin constituent which becomes possible [blending uniformly with thermoplastics the detailed inorganic filler which is easy to cause inorganic filler division secondary condensation by this invention], and discovers the outstanding mechanical property came to be obtained.

[Translation done.]

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TECHNICAL FIELD

[The technical field to which invention belongs] It distributes without an inorganic filler condensing in thermoplastics, and this invention has the outstanding mechanical and thermal property, and relates to the manufacture method at the resin constituent row which is excellent in the appearance of mold goods.

[Translation done.]

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PRIOR ART

[Description of the Prior Art] By adding an inorganic filler to thermoplastics, raising the intensity, rigidity, and thermal resistance is known well. However, when the particle diameter of the inorganic filler to be used is very small, particles condense, distribution in thermoplastics becomes poor, and there are problems -- shock resistance is spoiled. Moreover, differential powder of the sheet silicate is carried out to nano meter order into the polyamide which is thermoplastic engineering plastics, and the mechanical strength and the attempt which improves rigidity are performed in recent years. In this clay-polyamide complex, although rigidity is improved sharply, for that, a sheet silicate (clay) calls it about 1nm in thickness -- it is necessary to distribute in a polyamide in the state of a monolayer mostly. However, usually, a sheet silicate and thermoplastics were mixed and kneaded, secondary condensation arose in the request, and uniform distribution into a resin was difficult. Although examination which is going to obtain uniform distribution by using the intercalation compound which makes a sheet silicate a host at JP.8-12881,A, and makes specific quarternary ammonium salt a guest is made, improvement in much more mechanical property is called for. Moreover, the thing for which the vent-port which carried out melting kneading with the resin after making JP.8-151449,A and JP.9-48856,A swell a clay mineral with a solvent, and was prepared in the extruder is held to reduced pressure. Although the attempt which is going to obtain uniform distribution by removing a solvent was indicated, in order to use a solvent, when there was a problem that the process becomes complicated, the usual solvent had the problem that a thing with the operation which swells a sheet silicate did not have sufficient operation to thermoplastics, either.

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EFFECT OF THE INVENTION

[Effect of the Invention] The resin constituent which becomes possible [blending uniformly with thermoplastics the detailed inorganic filler which is easy to cause inorganic filler division secondary condensation by this invention], and discovers the outstanding mechanical property came to be obtained.

[Translation done.]

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] Then, this invention makes it a technical problem to obtain the resin constituent which makes it come uniformly to distribute, without making the small filler and small sheet silicate of solving an above-mentioned problem, i.e., an inorganic filler division particle diameter, condense in thermoplastics.

[Translation done.]

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MEANS

[Means for Solving the Problem] Then, the result which repeated examination wholeheartedly in order that this invention persons might solve an above-mentioned problem. By making low-molecular [low-molecular / of the super-critical state], i.e., supercritical fluid, act, in case thermoplastics is made to distribute the small filler and small sheet silicate of an inorganic filler division particle size. On the usual kneading conditions, the uniform distributed state which cannot be acquired because of secondary condensation could be attained, and it found out that the resin constituent which has the mechanical property which was excellent as the result was obtained, and resulted in this invention.

[0005] That is, this inventions are the resin constituent which (A) thermoplastics and (B) inorganic filler are made to contact (C) supercritical fluid, and comes to knead them, and its manufacture method.

[0006]

[Embodiments of the Invention] this invention is explained in detail below.

[0007] If (A) thermoplastics in this invention is a resin which can carry out plastic working by heating, either can be used regardless of amorphia, crystallinity, and mesomorphism. (A) As a desirable example of thermoplastics, polyester resin, polyamide resin, the poly arylene sulfide resin, polyacetal resin, ABS plastics, a polyphenylene-oxide resin, polycarbonate resin, polystyrene resin, polyolefin resin, acrylic resin, etc. are mentioned, and two or more sorts of these may be used together. Especially, polyester resin, polyamide resin, and the poly arylene sulfide resin can be used preferably.

[0008] With polyester resin here, the thermoplastic polyester which has a ring per chain of a polymer is mentioned, and the polymer or copolymer obtained by the condensation reaction which makes a principal component an aromatic dicarboxylic acid (or the ester plasticity derivative), a diol (or the ester plasticity derivative), and/or hydroxycarboxylic acid is usually specifically mentioned. Even if these are the things of mesomorphism, they may be the things of non-mesomorphism.

[0009] In this invention as an example of desirable polyester resin As a thing of non-mesomorphism, a polyethylene terephthalate, polypropylene terephthalate, To a polybutylene terephthalate and Pori, xylene terephthalate, polyethylene -2, 6-naphthalene dicarboxy rate, Others [rate / 4'-dicarboxy / a polybutylene -2, 6-naphthalene dicarboxy rate, polyethylene -1, 2-screw (phenoxy) ethane -4, and], Polyethylene isophthalate / terephthalate, polybutylene isophthalate / terephthalate, a polybutylene terephthalate / Deccan dicarboxy rate, polyethylene -4, a 4'-dicarboxy rate / terephthalate, etc. are mentioned.

[0010] The polyester which forms the anisotropy melting phase which consists of a structural unit chosen from the aromatic oxy-carbonyl unit, the aromatic dioxy unit, the aromatic dicarbonyl unit, the ethylene dioxy unit, etc. as a thing of mesomorphism can be mentioned.

[0011] As an aromatic oxy-carbonyl unit, for example as the structural unit generated from para hydroxybenzoic acid and the 6-hydroxy-2-naphthoic acid, and an aromatic dioxy unit for example, as the structural unit generated from 4, a 4'-dihydroxy biphenyl, hydroquinone, or t-butyl hydroquinone, and an aromatic dicarbonyl unit For example, as a terephthalic acid, an isophthalic acid, 2, the structural unit generated from 6-naphthalene dicarboxylic acid, and an aromatic imino OKISHI unit, the structural unit generated from 4-aminophenol is mentioned, for example.

[0012] It is also suitable practically to divide as polyester resin, and for a polybutylene terephthalate, a polyethylene terephthalate, polyethylene -2, and 6-naphthalene dicarboxy rate to be mentioned as a desirable thing, and to use these polyester resin as mixture according to required properties, such as a moldability, thermal resistance, toughness, and front-face nature.

[0013] Polyamide resin is a polyamide which uses amino acid, a lactam or a diamine, and a dicarboxylic acid as a main raw material. As an example of representation of the raw material, 6-aminocaproic acid, a 11-amino undecanoic acid, Amino acid, such as 12-amino dodecanoic acid and an paraamino methyl benzoic acid, Lactams, such as an epsilon caprolactam and omega-RAURO lactam, a tetramethylenediamine, A hexamethylenediamine, 2-methyl pentamethylene diamine, an undecamethylene diamine, A dodeca methylene diamine, 2 and 2, 4-/4, a 4-trimethyl hexamethylenediamine, [2 and 4] 5-methyl nonamethylene diamine, a meta-xylene diamine, a PARAKI silylene diamine, 1, 3-screw (aminomethyl) cyclohexane, 1, 4-screw (aminomethyl) cyclohexane, 1-amino-3-aminomethyl - 3, 5, and 5-trimethyl cyclohexane, Screw (4-amino cyclohexyl) methane, screw (3-methyl-4-amino cyclohexyl) methane, 2 and 2-screw (4-amino cyclohexyl) propane, a screw (aminopropyl) piperazine, Aliphatic series, such as an aminoethyl piperazine, an alieycle group, an aromatic diamine, And an adipic acid, a SUPERIN acid, an azelaic acid, a sebacic acid, dodecane diacid, A terephthalic acid, an isophthalic acid, 2-chloro terephthalic acid, 2-methyl terephthalic acid, 5-methyl isophthalic acid, 5-sodium sulfoisophtharate, a hexahydro terephthalic acid, Aliphatic series, such as a hexahydro isophthalic acid, an alieycle group, and an aromatic dicarboxylic acid are mentioned, and the polyamide homopolymer or copolymer guided from these raw materials can be respectively used in the form of independent or mixture in this invention.

[0014] In this invention especially useful polyamide resin It is polyamide resin excellent in the thermal resistance which has the melting point 200 degrees C or more, or intensity. As a concrete example, a PORIKA pro amide (nylon 6), a polyhexamethylene adipamide (Nylon 66), Polytetra ethylene adipamide (Nylon 46), a polyhexamethylene sebacamide (Nylon 610), Polyhexamethylene DODEKAMIDO (Nylon 612), a polyhexamethylene adipamide / polyhexamethylene terephthalamide copolymer (Nylon 66 / 6T), A polyhexamethylene adipamide / polyhexamethyleneisophthalamide copolymer (Nylon 66 / 6I), A polyhexamethylene adipamide / polyhexamethylene terephthalamide / polyhexamethyleneisophthalamide copolymer (Nylon 66 / 6T/6I), poly xylylene adipamide (nylon XD6) and such mixture, or a copolymer is mentioned. It is also suitable practically to be able to give examples, such as nylon 6, Nylon 66, Nylon 610, nylon 6/66 copolymer, and nylon 6/12 copolymer, and to use these Nylons as mixture as an especially desirable thing, according to required properties, such as a moldability, thermal resistance, toughness, and front-face nature, further.

[0015] There is no limit especially in the polymerization degree of these Nylons, and the thing of the range of 1.5-5.0, especially the range of 2.0-4.0 has the desirable relative viscosity measured at 25 degrees C among 1% of concentrated-sulfuric-acid solution.

[0016] As a poly arylene sulfide resin, it can use both a bridge formation type a straight chain type and branching type.

[0017] with (B) inorganic filler used for this invention, it is usually used as the filler of a resin, and a reinforcement -- fibrous or things [being un-fibrous] (for example, a grain, powder, or a tabular etc.) are pointed out As a desirable example of a fibrous inorganic filler, a glass fiber, a potassium titanate fiber, plaster fiber, brass fiber, stainless steel fiber, copper fiber, steel fiber, boron whisker fiber, a carbon fiber, etc. are mentioned. moreover, as a desirable example of the inorganic filler of a grain, powder, and a tabular A WARASUTE night, a sericite, a kaolin, a mica, clay, a bentonite, Silicate, such as talc and an alumina silicate, an alumina, silicon oxide, Metallic oxides, such as a magnesium oxide, a zirconium oxide, and titanium oxide, Carbonates, such as a calcium carbonate, a magnesium carbonate, and a dolomite, a calcium sulfate, Sulfates, such as a barium sulfate, a glass bead, a glass milled fiber, boron nitride, silicon carbide, Saroyan, etc. may be mentioned, and these may be hollow (for example, a hollow glass fiber, a glass micro balloon, a milt balloon, a carbon balloon, etc.). Although according to this invention the inorganic filler illustrated above is uniformly distributed in thermoplastics and the improvement effect of a mechanical property is accepted as a result, the effect of especially distributing the inorganic filler of a particle system uniformly is large, a thing with a fibrous particle system inorganic filler here -- the diameter -- about a grain and a powdery thing, the minor axis says [thing / of a tabular / the thickness] a thing 1 micrometer or less on an average As an inorganic filler with which the technology of this invention appears most notably, there is a silicate system inorganic filler and the effect over a sheet silicate is large also in especially inside. As an example of a sheet silicate, bloating tendency synthetic micas, such as various clay minerals, such as smectite system clay minerals, such as a montmorillonite, a beidellite, a nontronite, a saponite, a hectorite, a sauconite, and a vermiculite, a halloysite, a money dynamite, KENIYAITO, a zirconium phosphate, and phosphoric acid titanium, Li type fluorine TENIO light, Na type fluorine TENIO light, a Na type 4 silicon fluorine mica, and a Li type 4 silicon fluorine mica, etc. are mentioned, and it may be compounded even if it is a bloating tendency synthetic micas, such as smectite system clay minerals, such as a montmorillonite and a hectorite, and a Na type 4 silicon fluorine mica, Li type fluorine TENIO light, are desirable also in these. Although special processing is not necessarily needed when using a sheet silicate as a (B) inorganic filler in this invention, when a sheet silicate has the cation of convertibility between layers, the convertibility cation can use preferably the sheet silicate exchanged by the organic onium ion. In these, an ammonium ion and phosphonium ion are desirable and especially an ammonium ion is used preferably. As an ammonium ion, any of the 1st class ammonium, the 2nd class ammonium, the 3rd class ammonium, and the 4th class ammonium are sufficient.

[0018] As the 1st class ammonium ion, desyl ammonium, dodecyl ammonium, octadecyl ammonium, oleyl ammonium, benzyl ammonium, etc. are mentioned.

[0019] As the 2nd class ammonium ion, methyl dodecyl ammonium, methyl octadecyl ammonium, etc. are mentioned.

[0020] Dimethyl dodecyl ammonium, dimethyloctadecyl ammonium, etc. are mentioned as the 3rd class ammonium ion.

[0021] As the 4th class ammonium ion, benzyl trimethylammonium, Benzyl triethyl ammonium, benzyl tributyl ammonium, Benzyl trialkyl ammonium ions, such as benzyl dimethyl dodecyl ammonium and benzyl dimethyloctadecyl ammonium, Trioctyl methylammonium, trimethyl octyl ammonium, Alkyl trimethyl ammonium ions, such as trimethyl dodecyl ammonium and trimethyl octadecyl ammonium, Dimethyl dialkyl ammonium ions, such as dimethyl dioctyl ammonium, dimethyl didodecyl ammonium, and dimethyl dioctadecyl ammonium, etc. are mentioned.

[0022] Moreover, the ammonium ion guided besides these from an aniline, p-phenylene diamine, alpha-naphthylamine, a p-amino dimethylaniline, a benzidine, a pyridine, a piperidine, 6-aminocaproic acid, a 11-amino undecanoic acid, 12-amino dodecanoic acid, etc. is mentioned.

[0023] Also in these ammonium ions, the sum total of the carbon number in the molecule of an ammonium ion is suitable for especially the 4th class ammonium ion of 11-30. Specifically, they are trioctyl methylammonium, trimethyl octadecyl ammonium, benzyl dimethyl dodecyl ammonium, benzyl dimethyloctadecyl ammonium, etc.

[0024] Moreover, it is desirable to use the (B) inorganic bulking agent by the coupling agents (for example, an isocyanate system compound, an organic silane system compound, an organic titanate compound, an organic borane system compound, an epoxy compound, etc.) which have a reactant functional group, carrying out preliminary processing in the meaning which obtains the more excellent mechanical strength. Especially a desirable coupling agent is an organic silane system compound (silane coupling agent), as the example Gamma-glycidoxypropyltrimethoxysilane, gamma-glycidoxy propyl TORIETOKISHISHI silane, Epoxy-group content alkoxysilane compounds, such as beta-(3, 4-epoxycyclohexyl) ethyl trimethoxysilane, Sulfhydryl-group

content alkoxy silane compounds, such as gamma-mercapto propyltrimethoxysilane and gamma-mercapto propyl triethoxysilane, gamma-ureido propyl triethoxysilane, gamma-ureido propyl trimethoxy SHISHIRAN, Ureido machine content alkoxy silane compounds, such as gamma-(2-ureido ethyl) aminopropyl trimethoxysilane, gamma-isocyanato propyl triethoxysilane, gamma-isocyanato propyltrimethoxysilane, gamma-isocyanato propylmethyl dimethoxysilane, gamma-isocyanato propylmethyl diethoxysilane, gamma-isocyanatopropylethyl dimethoxysilane, gamma-isocyanatopropylethyl diethoxysilane, Isocyanato-group content alkoxy silane compounds, such as gamma-isocyanato propyl trichlorosilane, gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, Amino-group content alkoxy silane compounds, such as gamma-(2-aminoethyl) aminopropyl trimethoxysilane and gamma-aminopropyl trimethoxysilane, Hydroxyl-group content alkoxy silane compounds, such as gamma-hydroxypropyl trimethoxysilane and gamma-hydroxypropyl triethoxysilane, Carbon carbon unsaturation machine content alkoxy silane compounds, such as gamma-methacryloxypropyl trimethoxy silane, vinyltrimethoxysilane, and N-beta-(N-vinylbenzyl aminoethyl)-gamma-aminopropyl trimethoxysilane, a hydrochloride, etc. are mentioned. Especially, gamma-methacryloxypropyl trimethoxy silane, gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, gamma-(2-aminoethyl) aminopropyl trimethoxysilane, and gamma-aminopropyl trimethoxysilane are used preferably. Although the method of these silane coupling agents carrying out surface treatment of the (B) inorganic bulking agent beforehand according to a conventional method, and subsequently carrying out (A) thermoplastics and (C) melting kneading is used preferably, in case melting kneading of (A) thermoplastics and the (B) inorganic bulking agent is carried out under existence of low-molecular [of the (C) super-critical state], without performing surface treatment of an inorganic bulking agent beforehand, you may use the so-called integral blending method which adds these coupling agents.

[0025] Usually, although a coupling agent is used to an inorganic bulking agent in about 0.01 - 20% of the weight of the range, its 0.05 - 15 % of the weight is desirable.

[0026] As for (B) inorganic filler in this invention, it is desirable from the point of a moldability to blend preferably, 80 to 0.01% of the weight, by the inorganic ash content in a constituent, so that it may become 50 - 0.1 % of the weight. In addition, the inorganic ash content in a constituent is the value which was made to ash 2g of constituents for 3 hours, and asked for them with the 500-degree C electric furnace.